PB-245 406

RADIOCHEMICAL METHODOLOGY FOR DRINKING WATER

Herman L. Kreiger

Environmental Monitoring and Support Laboratory
Cincinnati, Ohio

September 1975

DISTRIBUTED BY:

NTIS
National Technical Information Service
U. S. DEPARTMENT OF COMMERCE

283203

EPA-600/4-75-008
September 1975

PB 243 806

Environmental Monitoring Series

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
US Department of Commerce
Springfield, VA. 22151

RESEARCH REPORTING SERIES

Research reports of the Office of Research and Development, U.S. Environmental Protection Agency, have been grouped into five series. These five broad categories were established to facilitate further development and application of environmental technology. Elimination of traditional grouping was consciously planned to foster technology transfer and a maximum interface in related fields. The five series are:

1. Environmental Health Effects Research
2. Environmental Protection Technology
3. Ecological Research
4. Environmental Monitoring
5. Socioeconomic Environmental Studies

This report has been assigned to the ENVIRONMENTAL MONITORING STUDIES series. This series describes research conducted to develop new or improved methods and instrumentation for the identification and quantification of environmental pollutants at the lowest conceivably significant concentrations. It also includes studies to determine the ambient concentrations of pollutants in the environment and/or the variance of pollutants as a function of time or meteorological factors.

This document is available to the public through the National Technical Information Service, Springfield, Virginia 22151.

TECHNICAL REPORT DATA

Please read Instructions on the reverse before completing

1. REPORT NO. EPA-600/4-75-008	2.
3. RECIPIENT'S ACCESSION NO. PB 245 106	
4. TITLE AND SUBTITLE: RADIOCHEMICAL METHODOLOGY FOR DRINKING WATER	5. REPORT DATE: September 1975 (Issuing Date)
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S): Herman L. Krieger	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Radiochemistry and Nuclear Engineering Branch, Environmental Monitoring and Support Laboratory, Cincinnati, Ohio 45268	10. PROGRAM ELEMENT NO. 2HBL20 (1HA327; ROAP 21-AAK; Task 005) 11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS: Office of Research and Development and Office of Radiation Programs, U.S. Environmental Protection Agency, Washington, D.C. 20460	13. TYPE OF REPORT AND PERIOD COVERED: In-house 14. SPONSORING AGENCY CODE: EPA-ORD

15. SUPPLEMENTARY NOTES

16. ABSTRACT

A laboratory manual of radiochemical procedures has been compiled and edited for use in the analysis of specific radionuclides in drinking water; nuclides for whom the Environmental Protection Agency recommended maximum contaminant levels in its interim drinking water standards. In addition to gross activity analyses, the procedures for $^{134}/^{137}Cs$, ^{131}I, $^{226}/^{228}Ra$, $^{89}/^{90}Sr$, ^{3}H and ^{40}K were evaluated by replicate testing to determine the method capabilities and minimum detection levels. The results, which indicate that the sensitivity of these procedures is at least a factor of ten greater than the present required limits, are given in the Appendix. Also appended is information on reagent preparation and suggested sources for purchasing special equipment.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Chemical analysis Potable water Quality control Radiactive contaminants Radiochemistry	Maximum contaminant level Minimum detection level Procedures Radiochemical analysis	13B

18. DISTRIBUTION STATEMENT: RELEASE TO PUBLIC	19. SECURITY CLASS (*This Report*): UNCLASSIFIED	21. NO. OF PAGES: 58
	20. SECURITY CLASS (*This page*): UNCLASSIFIED	22. PRICE: $3.25

EPA Form 2220-1 (9-73)

U.S. GOVERNMENT PRINTING OFFICE: 1975-657-695/5305 Region No. 5-11

EPA-600/4-75-008
September 1975

RADIOCHEMICAL METHODOLOGY FOR DRINKING WATER

by

Herman L. Krieger
Radiochemistry & Nuclear Engineering Branch
Environmental Monitoring and Support Laboratory
Cincinnati, Ohio 45268

Program Element No. 2FH120

Prepared for

Office of Radiation Programs
U.S. Environmental Protection Agency
Washington, D.C. 20460

ENVIRONMENTAL MONITORING AND SUPPORT LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
CINCINNATI, OHIO 45268

DISCLAIMER

This report has been reviewed by the Environmental Monitoring and Support Laboratory, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

FOREWORD

Environmental measurements are required to determine the quality of ambient waters and the character of waste effluents. The Environmental Monitoring and Support Laboratory - Cincinnati conducts research to:

- Develop and evaluate techniques to measure the presence and concentration of physical, chemical, and radiological pollutants in water, wastewater, bottom sediments, and solid waste.
- Investigate methods for the concentration, recovery, and identification of viruses, bacteria and other microbiological organisms in water. Conduct studies to determine the responses of aquatic organisms to water quality.
- Conduct an Agency-wide quality assurance program to assure standardization and quality control of systems for monitoring water and wastewater.

The Proposed Interim Drinking Water Regulations contain maximum contaminant levels for the concentration of drinking water. This manual which was developed by the staff of Radiochemistry Branch, EMSL, provides the methodology for monitoring the quality of drinking water for these radionuclides. In detail, it describes the analytical procedures that have been determined appropriate for measuring them with the necessary sensitivity and precision. This effort has been supported by the Office of Radiation Programs, U. S. Environmental Protection Agency, Washington, D. C.

Dwight G. Ballinger
Acting Director
Environmental Monitoring and Support Laboratory
Cincinnati

PREFACE

The Environmental Protection Agency (EPA), has recently published Proposed Interim Primary Drinking Water Regulations for radioactivity under the Safe Drinking Water Act, PL 93-523.[1] These regulations propose maximum contaminant levels limiting the concentrations of natural and man-made radioactivity in drinking water supplies and set out the proposed monitoring requirements. Subsequent revisions of the proposed regulations may be made as pertinent information becomes available. However, the present Proposed Regulations set the following maximum contaminant levels for drinking water:

Radionuclides	Maximum Contaminant Level, pCi/l
Natural alpha particle activity ^{226}Ra + ^{228}Ra	5
Gross alpha activity (including ^{226}Ra)	15
Man-made radionuclides	a concentration not to produce an annual dose equivalent greater than 4 mrem per year.

The regulations specify the following detection limits for radionuclides of primary interest:

Gross alpha activity	1 pCi/l	Strontium-90	1/2 pCi/l
Radium-226	1 pCi/l	Iodine-131	1 pCi/l
Radium-228	1 pCi/l	Cesium-134	10 pCi/l
Tritium	1000 pCi/l	Gross Beta	10 pCi/l
Strontium-89	10 pCi/l		

where the detection limit is defined as that concentration that can be measured with an accuracy of $\pm$ 100% at the 95% confidence level, i.e., 2 sigma. Furthermore, the regulations require a radiochemical analysis if the gross beta activity exceeds 50 pCi per liter.

The Proposed Interim Regulations also recommend which methods may be applicable for the analyses, selecting them from many sources. Recognizing the need for a collection of methods that can be used for measuring each of these nuclides in drinking water, the Radiochemistry and Nuclear Engineering Branch of the Environmental Monitoring and Support Laboratory, EPA, has sought to bring various methods together in this one volume. Such a collection of sensitive procedures would be of significant value in making a dose assessment, and would serve as a source for less-experienced laboratories to improve their technical capabilities.

This compilation includes single-operator-tested procedures that have the required sensitivity for drinking water guidelines. They have been

selected from a number of radiochemical methodology collections,[2-6] and have been prepared so they are readily adaptable for routine analysis. Several of them have already been published as standard reference methods. Although specific sample volumes are designated in each of these procedures, most of the methods can easily be adapted for larger volumes with minor changes in the initial treatment. With such modifications, the minimum detection level (MDL) can be lowered two- or three-fold. Factors considered in selecting these procedures for this laboratory manual were procedure time and method capabilities, and only when modifications were deemed essential were the procedures lengthened to include scavenging or other purification steps. (See Appendix A.)

Drinking water is normally collected at "grab" sampling points and should not be acidified until the start of the analysis, if so indicated. Radiochemical analysis should be performed immediately upon receipt of sample, especially for ^{131}I so that radioactive decay losses are minimized. When quarterly composites are set aside for future analyses, the samples should be acidified with 1 ml 16 $\underline{N}$ HNO_3 per liter to minimize losses caused by adsorption on container wall. Preparation of reagents is described in Appendix B, and sources of supply in Appendix C.

Other methods effective for the task and the specifications required obviously are available or may become available in the future. Revisions or additions to these procedures are solicited and will be considered in a subsequent edition of the manual.

References:

1. Proposed Interim Primary Drinking Water Regulations - Radioactivity. U. S. Environmental Protection Agency (40 CFR Part 141). Notice of Proposed Maximum Contaminant Levels for Radioactivity. Federal Register, August 1975.

2. Krieger, H. L. and S. Gold. Procedures for Radiochemical Analysis of Nuclear Reactor Aqueous Solutions. EPA-R4-73-014, National Environmental Research Center, U. S. Environmental Protection Agency, Cincinnati, Ohio (May 1973).

3. 1972 Book of ASTM Standards, Part 23. American Society for Testing and Materials, Philadelphia, Pa. (1972).

4. Harley, J. H. Manual of Standard Procedures. USAEC Rept. HASL-300 (1972).

5. Nuclear Science Series, USAEC Rept. NAS-NS-3001 to NAS-NS-3111 (1965).

6. Standard Methods for the Examination of Water and Waste Water, 13th ed. American Public Health Association, Washington, D. C. (1971).

CONTENTS

	Page
Foreword	iii
Preface	iv
Figures	viii
Acknowledgment	ix
Gross Alpha and Beta Radioactivity in Drinking Water	1
Radioactive Cesium in Drinking Water	4
Radioactive Iodine in Drinking Water	
Precipitation Method	6
Distillation Method	9
Radium-226 in Drinking Water	
Precipitation Method	13
Radon Emanation Technique	16
Radium-228 in Drinking Water	
Sequential Method Radium-228/Radium-226	24
Radium-226 by Precipitation	26
Radium-226 by Radon Emanation	27
Radioactive Strontium in Drinking Water	29
Tritium in Drinking Water	34
Potassium-40 in Drinking Water	38
Appendices	
A. Method Capabilities	41
B. Reagant Preparation	42
C. Sources of Supply	47

FIGURES

		Page
1.	Generalized Gross Alpha and Gross Beta Self-Absorption Curves . . .	2
2.	Distillation Apparatus for Iodine Analysis	11
3.	Radon Emanation Apparatus with Scintillation Cell	17
4.	A Typical Radon Bubbler	18
5.	The Growth of Radon-222 from Radium-226	20
6.	A Typical Scintillation Cell for Radon Counting	21
7.	Distillation Apparatus for Tritium Analysis	35

ACKNOWLEDGMENT

The Radiochemistry & Nuclear Engineering Branch (R&NEB) of the Environmental Monitoring and Support Laboratory, EPA, was requested to prepare this manual by the end of fiscal year 75. It is one task milestone in the Research Objective Achievement Plan of the Office of Research and Development, and is intended for use by agencies responsible for monitoring drinking water for radioactive contamination. George W. Frishkorn, Mrs. Betty Jacobs, and Mrs. Eleanor Martin, R&NEB, were responsible for the thorough testing of these procedures and for recommending modifications so that they satisfy the limiting concentrations of the standard. Editorial comments and suggestions for improvement in the clarity of the manual were submitted by Dr. Bernd Kahn, Environmental Resources Center, Georgia Institute of Technology, Atlanta, Georgia and Mr. Robert Lieberman, Eastern Environmental Radiation Laboratory, Montgomery, Alabama.

Their assistance is gratefully acknowledged.

GROSS ALPHA AND BETA RADIOACTIVITY IN DRINKING WATER

Principle of Method

A specified volume of drinking water is evaporated to dryness, transferred to a counting dish, and counted for gross alpha or gross beta activity.

Procedure Time

Evaporation time, 1 to 2 hours for eight 250-ml samples.

Reagents and Supplies

Planchets, stainless steel.

Procedure

1. Transfer 100 to 500 ml drinking water sample (Note 1) to a beaker and evaporate to dryness on a hot plate. Do not bake.
2. Slurry residue to a tared stainless-steel planchet using a rubber policeman and as little water as possible.
3. Dry under infra-red lamps, cool, weigh, and store in desiccator.
4. Alpha and beta count (Note 2).

Calculation

Calculate the concentration, D, of the gross activity (alpha and/or beta) in picocuries per liter as follows:

$$D = \frac{C \times 10^3}{2.22 \times EVW}$$

where:

C = net count rate, counts/minute,
E = counter efficiency, alpha or beta (Note 3),
V = milliliters of sample used,
W = self-absorption factor (determined from solids concentration), and
2.22 = conversion factor from disintegrations/minute to picocuries.

Notes:

1. The specified volume of drinking water to be evaporated will be a function of its hardness and solids concentration. Self-absorption factors for the solids present in these volumes have to be determined to correct for losses due to self-absorption. In any event, the maximum sample thickness should be less than 3 mg/cm^2. (See Fig. 1.)

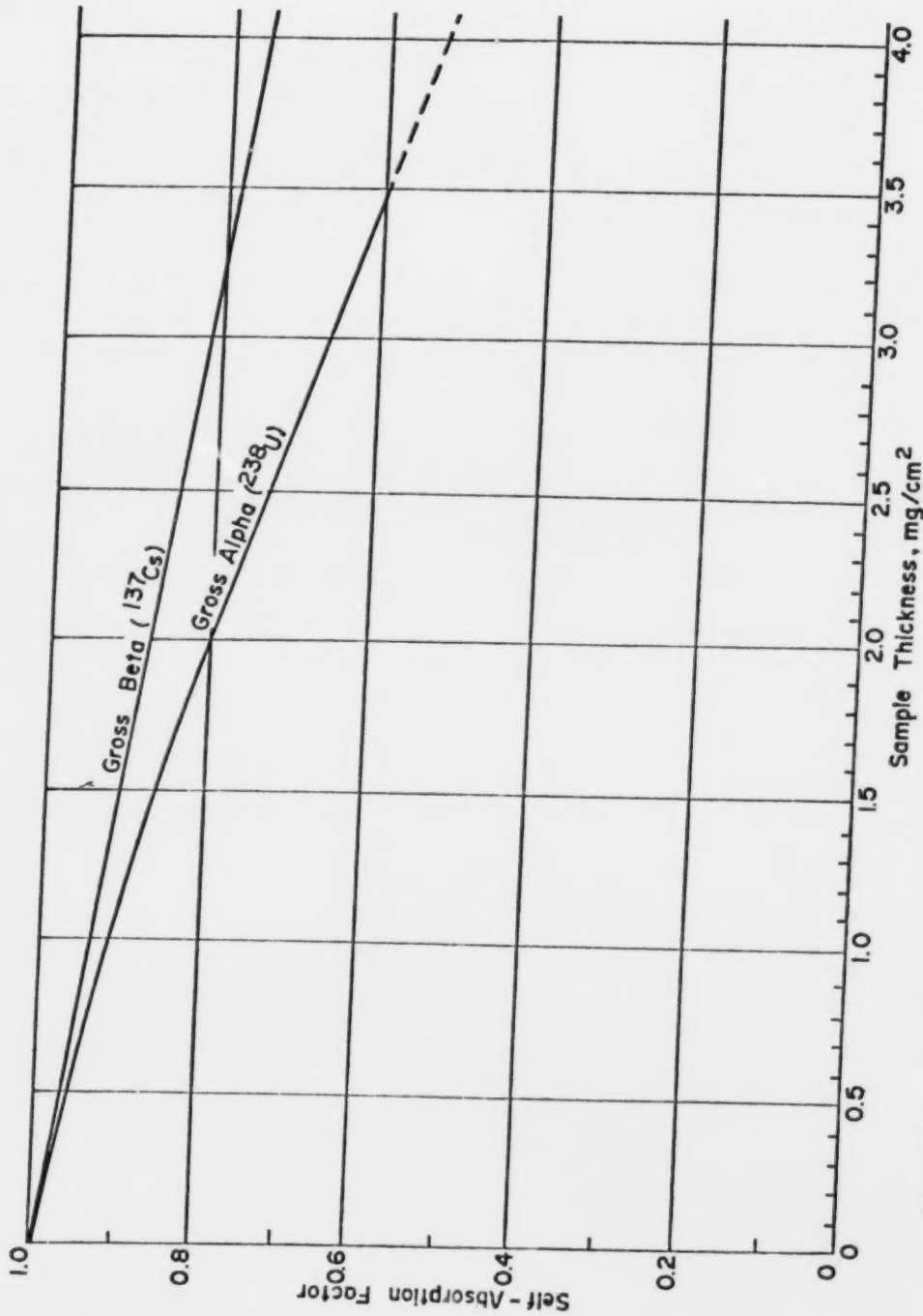

Figure 1. Generalized Gross Alpha and Gross Beta Self-Absorption Curves

2. The same planchet can be counted for alpha and beta activities in designated instruments provided their counting chambers are capable of handling the same size planchet.

3. It is recommended that NBS-calibrated standards be used for ascertaining instrument efficiencies. A weightless deposit of ^{210}Po for α and a point source from a standard solution of ^{137}Cs for β are suggested. Source of supply is: Standard Reference Materials Catalog, NBS Publication 260, U. S. Department of Commerce (1974). Standards should also be prepared in the geometry and weight ranges to be encountered in these gross analyses.

Literature

Standard Methods for the Examination of Water and Waste Water, 13th ed. American Public Health Association, Washington, D. C. (1971).

RADIOACTIVE CESIUM IN DRINKING WATER

Principle of Method

Cesium carrier is added to the aqueous sample. The cesium is collected as the phosphomolybdate and purified as Cs_2PtCl_6 for counting.

Procedure Time

Four samples in 5 hours.

Reagents

Ammonium phosphomolybdate, $(NH_4)_3PMo_{12}O_{40}$: prepared reagent
Calcium chloride, $CaCl_2$: 3 M
Cesium carrier: 10 mg/ml
Chloroplatinic acid, $H_2PtCl_6 \cdot 6H_2O$: 0.1 M
Ethanol, C_2H_5OH: 95%
Hydrochloric acid, HCl: 12 N (conc.), 6 N, 1 N
Sodium hydroxide, NaOH: 6 N

Procedure

1. To a 1000-ml drinking water sample, add 1.0 ml cesium carrier and enough 12 N HCl to make the solution ~0.1 N HCl.
2. Slowly add 1 gram $(NH_4)_3PMo_{12}O_{40}$ and stir for 30 minutes, using a magnetic stirrer. Allow precipitate to settle for at least 4 hours, and discard supernatant.
3. Slurry precipitate into a centrifuge tube. Centrifuge and discard supernatant.
4. Wash precipitate with 20 ml 1 N HCl, and discard wash solution.
5. Dissolve precipitate by dropwise addition of 3 to 5 ml 6 N NaOH. Heat over a flame for several minutes to remove ammonium ions. (Moist pH paper turns green as long as NH_3 vapors are evolved.) Dilute to 20 ml with water.
6. Add 10 ml 3 M $CaCl_2$ and adjust to pH 7 with 6 N HCl to precipitate $CaMoO_4$. Stir, centrifuge, and filter supernatant through Whatman #41 filter paper (or equivalent) into a 50-ml centrifuge tube.
7. Wash the precipitate remaining in the centrifuge tube with 10 ml water; filter through the same filter paper, and combine the wash with filtrate.

Discard filter paper.

8. Add 2 ml 0.1 M H_2PtCl_6 and 5 ml ethanol. Cool and stir in ice bath for 10 minutes.
9. Transfer with water to a tared glass-fiber filter. Wash with successive portions of water, 1 N HCl, and ethanol.
10. Dry, cool, weigh, mount, and beta count.

Calculation

Calculate the concentration, D, of the cesium activity in picocuries per liter as follows:

$$D = \frac{C}{2.22 \times EVR}$$

where:

C = net count rate, counts/minute,
E = counter efficiency,
V = liters of sample used,
R = fractional chemical yield, and
2.22 = conversion factor from disintegrations/minute to picocuries.

Literature:

Finston, H. L., and M. T. Kinsley. The Radiochemistry of Cesium. AEC Rept. NAS-NS-3035 (1961).

RADIOACTIVE IODINE IN DRINKING WATER

Precipitation Method

Principle of Method

Iodate carrier is added to an acidified sample of drinking water and, after reduction with Na_2SO_3 to iodide, the ^{131}I is precipitated with $AgNO_3$. The precipitate is dissolved and purified with zinc powder and sulfuric acid, and the solution is reprecipitated as PdI_2 for counting.

Procedure Time

Eight samples in 6 hours.

Reagents

Ammonium hydroxide, NH_4OH: 6 N
Ethanol, C_2H_5OH: 95%
Hydrochloric acid, HCl: 6 N
Iodate carrier, (IO_3^-): 10 mg/ml
Nitric acid, HNO_3: 16 N (conc.)
Palladium chloride, $PdCl_2$: 0.2 M
Silver nitrate, $AgNO_3$: 0.1 M
Sodium sulfite, Na_2SO_3: 1 M (freshly prepared)
Sulfuric acid, H_2SO_4: 2 N
Zinc, powder: reagent grade

Procedure

1. To a 2000-ml drinking water sample, add 15 ml 16 N HNO_3 and 1.0 ml iodate (IO_3^-) carrier. Mix well.
2. Add 4 ml freshly prepared 1 M Na_2SO_3 and stir for 30 minutes.
3. Add 20 ml 0.1 M $AgNO_3$, stir for 1 hour and settle for another hour.
4. Decant and discard as much as possible of the supernatant. Filter the remainder through a glass-fiber filter and discard filtrate.
5. Transfer the filter to a centrifuge tube and slurry with 10 ml water. Add 1 gram zinc powder and 2 ml 2 N H_2SO_4, and stir frequently for at least 30 minutes.
6. Filter, with vacuum, through a fine-fritted glass funnel and collect filtrate in an Erlenmeyer flask. Use a few ml water to wash both residue and filter and add the wash to filtrate in the flask. Discard residue.
7. Add 2 ml 6 N HCl to the filtrate and heat. Add 1 ml 0.2 M $PdCl_2$ and

digest for at least 5 minutes. Centrifuge and discard supernatant.

8. Dissolve the precipitate in 5 ml 6 N NH_4OH and heat in boiling water bath. Filter through a glass-fiber filter and collect filtrate in a centrifuge tube. Discard filter and residue.
9. Neutralize the filtrate with 6 N HCl, add 2 ml additional, and heat in a water bath. Add 1 ml 0.2 M $PdCl_2$ to reprecipitate PdI_2 and digest for 10 minutes. Cool slightly.
10. Transfer to a tared glass-fiber filter with water (Note 1). Wash successively with 5 ml portions of water and ethanol.
11. Dry to constant weight at $< 100^{o}C$, mount, and beta count (Note 2).

Calculation

Calculate the concentration, D, of the iodine activity in picocuries per liter as follows:

$$D = \frac{C}{2.22 \times EVR \times A}$$

where:

C = net count rate, counts/minute (c/m),
E = counter efficiency,
V = liters of sample used,
R = fractional chemical yield,
A = decay correction for ^{131}I ($t_{1/2}$ = 8.06 d), and
2.22 = conversion factor from disintegrations/minute to picocuries.

Notes:

1. Ordinarily, the final PdI_2 precipitate is collected on a glass-fiber filter and counted in a G-M system where the natural background is about 0.8 c/m. Glass-fiber filters add about 0.5 c/m to the background so that the PdI_2 counted this way will have about a 1.3 c/m background and a counting efficiency of 35%. However, if the precipitate is collected on a 0.8-μ membrane filter, and dried for 30 minutes at $70^{o}C$, it is possible to count the PdI_2 in a beta-gamma coincidence scintillation system where the background coincidence count is less than 0.1 c/m and the counting efficiency is greater than 40% (see Brauer *et al.* reference).

2. Confirmation of half-life of ^{131}I and indication of purity of the analysis can be made by recounting the planchet in approximately 1 week.

Literature:

Brauer, F. P., J. H. Kaye, and R. E. Connaly. X-ray and β-γ Coincidence Spectrometry Applied to Radiochemical Analysis of Environmental Samples. Advances in Chemistry Series, No. 93, Radionuclides in the

Environment, pp. 231-253. American Chemical Society (1970).

Kleinberg, J., and G. A. Cowan. The Radiochemistry of Fluorine, Chlorine, Bromine and Iodine. AEC Rept. NAS-NS-3005 (1960).

RADIOACTIVE IODINE IN DRINKING WATER

Distillation Method

Principle of Method

Iodide carrier is added to a drinking water sample, which is acidified to effect distillation of iodine into caustic solution. The distillate is acidified and the iodine is extracted into CCl_4. After back extraction, the iodine is purified as AgI or PdI_2 for counting.

Procedure Time

Two samples in 6 hours.

Reagents

Ammonium hydroxide, NH_4OH: 15 N (conc.)
Carbon tetrachloride, CCl_4
Diethyl ether, $(C_2H_5)_2O$: anhydrous
Ethanol, C_2H_5OH: 95%
Iodide carrier: 20 mg/ml
Nitric acid, HNO_3: 16 N (conc.), 4 N, 0.2 N
Silver nitrate, $AgNO_3$: 0.1 M
Sodium bisulfite, $NaHSO_3$: 1 M
Sodium hydroxide, NaOH: 0.5 N
Sodium nitrite, $NaNO_2$: 1 M
Sulfuric acid, H_2SO_4: 12 N
Tartaric acid, $C_4H_6O_6$: 50%

Procedure

1. To a 2000-ml drinking water sample in a 3-liter round-bottom flask, add 15 ml 50% $C_4H_6O_6$ and 1.0 ml iodide carrier. Mix well, cautiously add 25 ml cold 16 N HNO_3, and close the distillation apparatus (Fig. 2) (Note 1).
2. Connect an air line to the inlet, adjust the flow rate to about 2 bubbles/second, then distill for at least 15 minutes into 15 ml 0.5 N NaOH. Cool and transfer the NaOH solution to a 60-ml separatory funnel. Discard the solution in the round-bottom flask.
3. Adjust the solution to slightly acid with 1 ml 12 N H_2SO_4 and oxidize with 1 ml 1 M $NaNO_2$. Add 10 ml CCl_4 and shake for 1 to 2 minutes. Draw off organic layer into a clean 60-ml separatory funnel containing 2 ml 1 M $NaHSO_3$.
4. Add 5 ml CCl_4 and 1 ml 1 M $NaNO_2$ to the original separatory funnel

containing the aqueous layer and shake for 2 minutes. Combine the organic fraction with that in the separatory funnel in step 3.

5. Repeat step 4 and discard the aqueous layer.
6. Shake separatory funnel thoroughly until CCl_4 layer is decolorized; allow phases to separate and transfer aqueous layer to a centrifuge tube.
7. Add 2 ml 1 M $NaHSO_3$ to separatory funnel which has the CCl_4 and shake for several minutes. When the phases separate, combine this aqueous layer with that in centrifuge tube from step 6.
8. Add 1 ml water to separatory funnel and shake for several minutes. When the phases separate, add aqueous layer to the same centrifuge tube. Discard organic layer. (Note 2).
9. To the combined aqueous fractions, add 2 ml 0.1 M $AgNO_3$ plus 4 ml HNO_3. Stir for 1 minute then allow to stand 30 minutes at room temperature to coagulate silver iodide. Centrifuge at high speed for 10 minutes. Carefully decant and discard supernatant.
10. Pipet 5 ml 15 N NH_4OH into the centrifuge tube and stir thoroughly for several minutes. Heat carefully while stirring until boiling begins. Centrifuge at high speed for 10 minutes. Carefully decant and discard supernatant.
11. Slurry precipitate in 10 ml water, add 1 ml 4 N HNO_3, stir and immediately transfer precipitate to tared glass-fiber filter. Filter slowly at first to prevent loss.
12. Wash precipitate on filter successively with 20 ml 0.2 N HNO_3, 20 ml ethanol and 20 ml diethyl ether. Let dry with suction at least 10 minutes. Store in desiccator.
13. Weigh, mount, and store in light-tight container until ready to beta count.

Calculation

Calculate the concentration, D, of the iodine activity in picocuries per liter as follows:

$$D = \frac{C}{2.22 \times EVR \times A}$$

where:

C = net count rate, counts/minute,

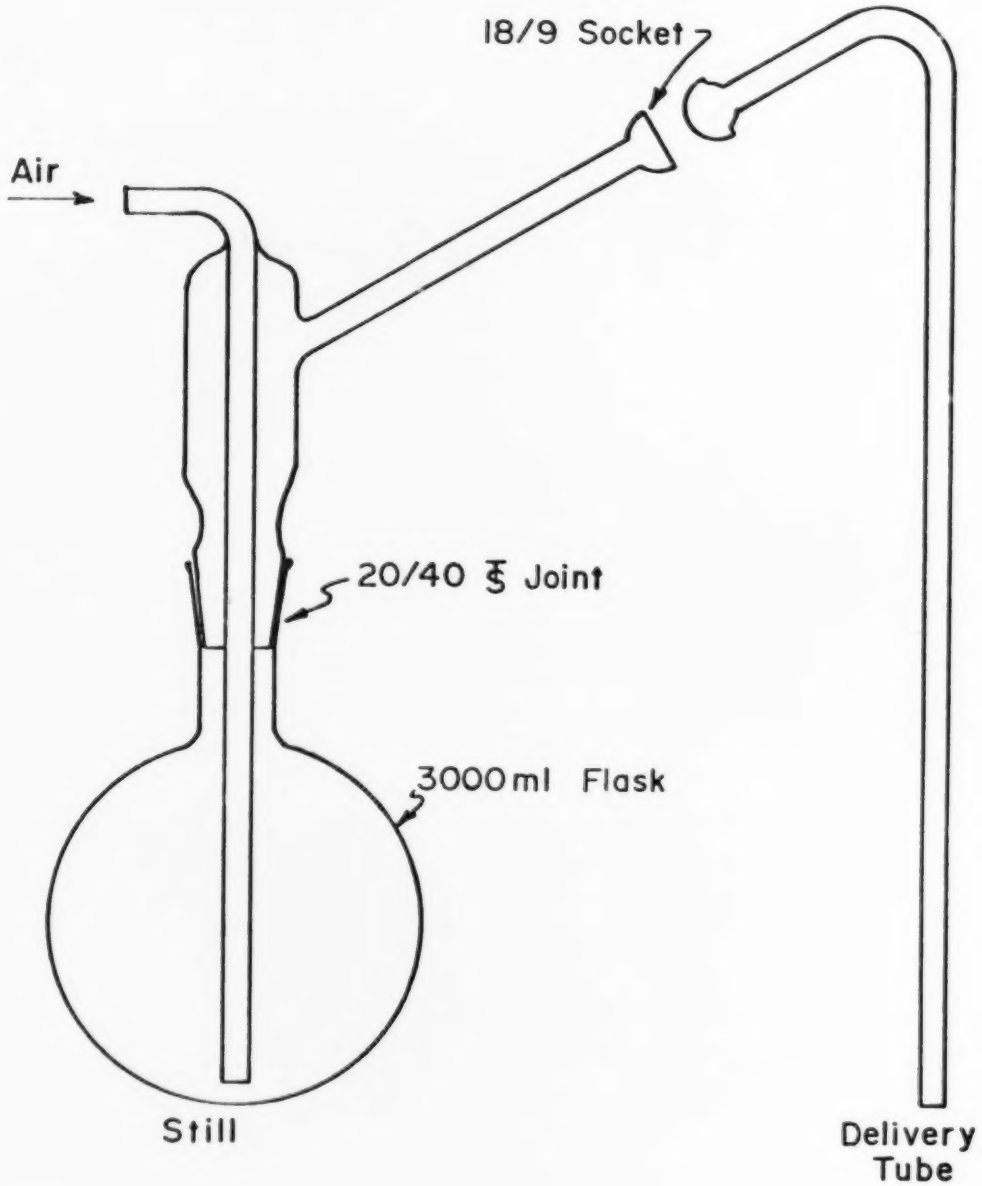

Figure 2. Distillation Apparatus for Iodine Analysis

E = counter efficiency,
V = liters of sample used,
R = fractional chemical yield,
A = decay correction for ^{131}I ($t_{1/2}$ = 8.06 d), and
2.22 = conversion factor from disintegrations/minute to picocuries.

Notes.

1. A distillation apparatus such as shown in Figure 2 or equivalent can be used: a round-bottom flask fitted with a ground-glass joint that allows for a source of air to be bubbled into the sample and that has a delivery tube on the other end extending into a caustic trap.

2. An alternate method, whereby the iodide is precipitated as PdI_2, provides a means for measuring radioiodine with less interference from chlorides.

   The procedure, after step 8, would then be:

   a. To the combined aqueous fractions, add 2 ml 6 N HCl and heat. Add 2 ml 0.2 M $PdCl_2$ and digest for at least 5 minutes. Centrifuge and discard supernatant.

   b. Transfer to a tared glass-fiber filter with water. Wash twice with 5-ml portions of water and ethanol.

   c. Dry to constant weight at 70 to 90°C, mount, and beta count.

Literature:

1972 Book of ASTM Standards, Part 23. American Society for Testing and Materials, Philadelphia, Pa. D 2334-68 (1972).

RADIUM-226 IN DRINKING WATER

Precipitation Method

Principle of Method

The radium in the drinking water sample is coprecipitated with barium as the sulfate and purified by reprecipitation from EDTA solution. The $BaSO_4$ precipitate containing ^{226}Ra is counted for alpha activity.

Procedure Time

One sample in 6 hours; four samples in 8 hours.

Reagents

Acetic acid, CH_3COOH: 17.4 N (glacial)
Ammonium hydroxide, NH_4OH: 15 N (conc.), 2 N
Barium carrier: 16 mg/ml
Citric acid, $C_6H_8O_7.H_2O$: 1 M
EDTA reagent: prepared reagent
Indicator, methyl orange: 0.1%
Lead carrier: 15 mg/ml
Sulfuric acid, H_2SO_4: 18 N
Sodium hydroxide, NaOH: 6 N

Procedure

1. To a 1000-ml drinking water sample, add 5 ml 1 M $C_6H_8O_7.H_2O$, 2.5 ml 15 N NH_4OH, 1 ml lead carrier, and 2.0 ml barium carrier.
2. Heat the solution to boiling and add 10 drops of methyl orange indicator.
3. Add, with stirring, 18 N H_2SO_4 to a permanent pink color and then 0.5 ml in excess.
4. Digest 5 to 10 minutes and let the mixed $BaSO_4$-$PbSO_4$ precipitate settle overnight. Decant and discard supernatant.
5. Transfer the precipitate to a centrifuge tube with a minimum amount of water. Centrifuge and discard supernatant.
6. Dissolve the precipitate by adding 15 ml EDTA reagent; heat in a water bath until clear. If not entirely clear, continue heating and add a few drops of 6 N NaOH.
7. Add 2 ml 17.4 N CH_3COOH dropwise and digest 5 to 10 minutes (Note 1). Centrifuge, discard the supernatant, and record time (Note 2).
8. Wash the $BaSO_4$ precipitate containing the ^{226}Ra with 15 ml 2 N NH_4OH. Centrifuge and discard wash.

9. Wash the $BaSO_4$ precipitate with 10 ml water, centrifuge, and discard wash.
10. Transfer the precipitate to a tared stainless-steel planchet with a minimum of water, and dry under infra-red lamps (Note 3).
11. Cool, weigh, and store in desiccator.
12. Determine the alpha activity in an internal proportional counter (Note 4).

Calculation

Calculate the concentration, D, of the ^{226}Ra activity (which would include any ^{224}Ra and ^{223}Ra that is present) in picocuries per liter as follows:

$$D = \frac{C}{2.22 \times EVR \times IW}$$

where:

C = net count rate, counts/minute,
E = counter efficiency for alpha counting,
V = liters of sample used,
R = fractional chemical yield,
I = ingrowth factor (Note 5),
W = self-absorption factor, and
2.22 = conversion factor from disintegrations/minute to picocuries.

Notes:

1. This volume of acetic acid gives a pH of about 4.5 and is sufficient to destroy the Ba-EDTA, but not the Pb-EDTA, complex.
2. At this step of the procedure, radon (and daughters) grows into the $BaSO_4$ precipitate.
3. Drying should be rapid but not too vigorous to minimize loss of ^{222}Rn that has already grown into the precipitate.
4. Alpha self-absorption losses for 226Ra in an internal proportional counter as a function of sample thickness are:

Sample thickness, mg/cm^2	Relative efficiency losses, %
0.0	0.00
0.31	0.04
0.95	0.06
1.26	0.12
1.55	0.14
1.90	0.19
1.97	0.20
2.20	0.22
2.65	0.30
3.14	0.37

5. The ingrowth factor can be calculated from Kirby's data (see Kirby, H. W. reference). Some of these alpha activities from initially pure ^{226}Ra during the first 240 hours are:

Hours	Ingrowth factor
0	1.0000
1	1.0160
2	1.0363
3	1.0580
4	1.0798
5	1.1021
6	1.1238
24	1.4892
48	1.9054
72	2.2525
96	2.5422
120	2.7838
144	2.9853
192	3.2939
240	3.5086

Literature:

Goldin, A. S. Determination of Dissolved Radium. Anal. Chem. 33, 406-409 (March 1961).

Harley, J. H., ed. Manual of Standard Procedures, USAEC Rept. HASL 300 (1972).

Kirby, H. W. Decay and Growth Tables for the Naturally Occurring Radioactive Series. Anal. Chem. 26, 1063-1071 (1954).

Although this procedure utilizes an internal proportional counter to determine alpha activity, it can be modified so that the final precipitate (step 9) is filtered on tared Whatman #42 filter paper, dried, weighed, and covered with an alpha phosphor and Mylar. After storing for 30 days, the ^{226}Ra in the sample can be determined with an alpha scintillation counter.

RADIUM-226 IN DRINKING WATER

Radon Emanation Technique

Principle of Method

The radium in the drinking water sample is concentrated and separated by coprecipitation with barium as the sulfate. The precipitate is dissolved in EDTA reagents, placed in a sealed bubbler and stored for ingrowth of ^{222}Rn.

Procedure Time

Four samples in 8 hours.

Equipment

Radon Emanation Apparatus with Scintillation Cell (Fig. 3).

Reagents

Ammonium hydroxide, NH_4OH: 15 N (conc.)
Ascarite: drying reagent, 8-20 mesh
Barium carrier: 16 mg/ml
EDTA reagent: 0.25 M
Helium gas
Hydrochloric acid, HCl: 12 N (conc.)
Magnesium perchlorate, $Mg(ClO_4)_2$: reagent grade
Sulfuric acid, H_2SO_4: 18 N, 0.1 N

Procedure

1. To a 1000-ml drinking water sample, add 20 ml 12 N HCl and 2.0 ml barium carrier and heat to boiling.
2. Cautiously and with vigorous stirring, add 20 ml 18 N H_2SO_4. Digest 5 to 10 minutes and let precipitate settle overnight. Decant and discard supernatant.
3. Slurry the precipitate and transfer to a centrifuge tube with a minimum amount of 0.1 N H_2SO_4. Centrifuge and discard supernatant. Wash twice with 0.1 N H_2SO_4 and discard washes.
4. Add 20 ml EDTA reagent, 2 drops 15 N NH_4OH and heat in a water bath until the precipitate dissolves (Note 1).
5. Transfer the solution to a radon bubbler (Fig. 4). Open both the upper and lower stopcocks and de-emanate the solution by slowly passing helium gas through the bubbler for about 20 minutes.

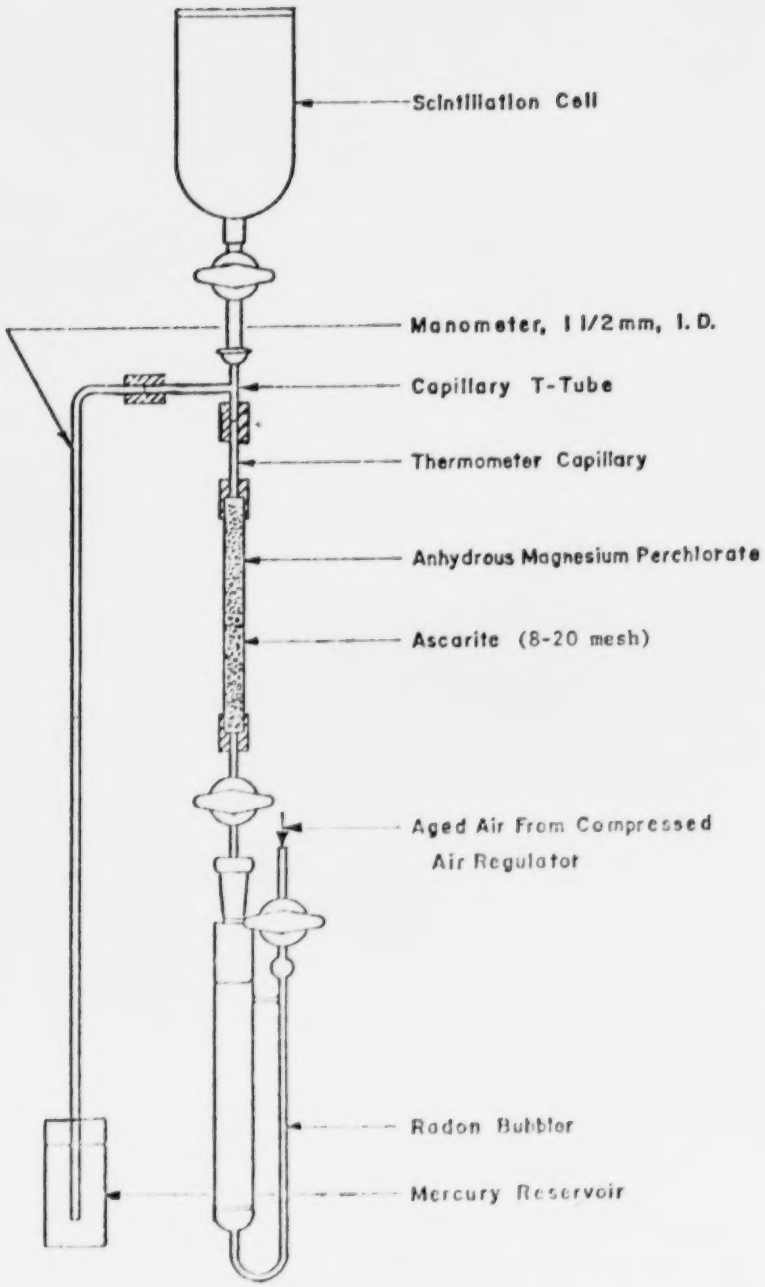

Figure 3. Radon Emanation Apparatus with Scintillation Cell

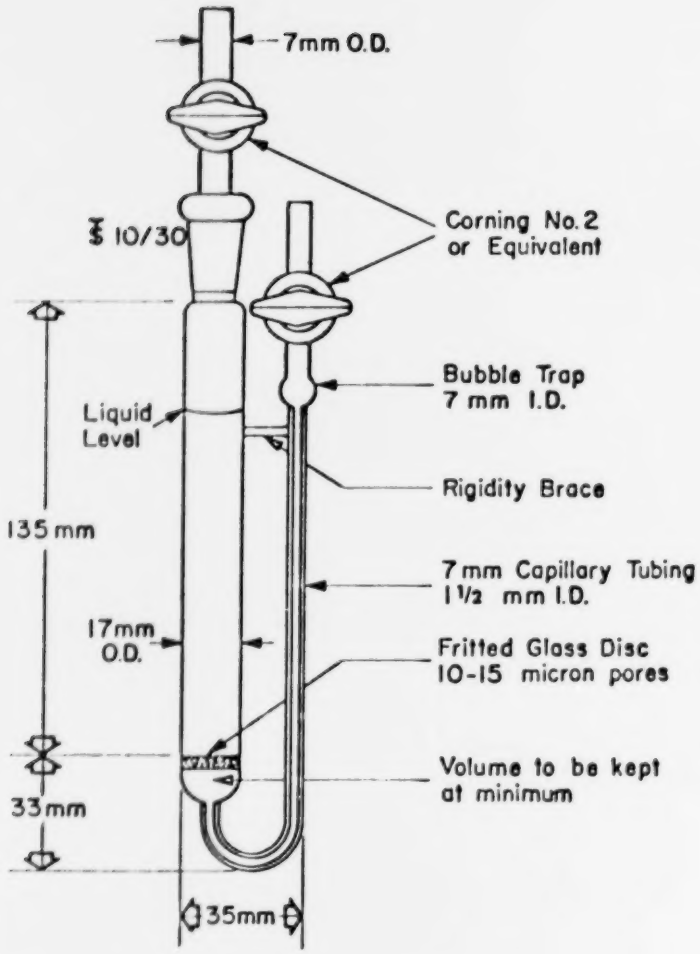

Figure 4. A Typical Radon Bubbler (emanation tube)

6. Close the two stopcocks, and record time. Store the solution for 4 to 8 days for ingrowth of ^{222}Rn (Fig. 5).
7. At the end of the storage period, fill the upper half of an absorption tube with magnesium perchlorate and the lower half with Ascarite (Note 2). Attach the tube to the radon bubbler and then attach the evacuated scintillation cell (Fig. 6) to the tube.
8. Open the stopcock on the cell and check the assembly for leaks. Gradually open the outlet stopcock on the bubbler, and when the stopcock is fully open and no further significant bubbling takes place, close the stopcock.
9. Adjust the helium gas pressure so that the gas flows at slightly above atmospheric pressure.
10. Connect the hose to the bubbler inlet and gradually open the inlet stopcock using the bubbling as a guide. When the stopcock can be fully opened without a significant amount of bubbling, the bubbler is essentially at atmospheric pressure again.
11. Open the outlet stopcock very slightly and allow bubbling to proceed at a rate, determined by experience, such that 15 to 20 minutes are required to complete de-emanation.
12. Toward the end of the de-emanation, when the vacuum is no longer effective, gradually increase the helium gas pressure. When the system is at atmospheric pressure, shut off the helium gas, disconnect the tubing from the bubbler inlet and close the inlet and outlet stopcocks of the cell and bubbler, and record time. This the beginning of ^{222}Rn decay and ingrowth of ^{222}Rn daughters.
13. Store the scintillation cell for at least 4 hours to ensure equilibrium between radon and radon daughters. Count the alpha scintillations from the cell in a radon counter with a light-tight enclosure that protects the photomultiplier tube. Record the counting time to correct for the decay of ^{222}Rn (Note 3).

Calculation

Calculate the concentration, D, of the ^{226}Ra activity in picocuries per liter as follows:

$$D = \frac{C}{2.22\ EV} \times \frac{1}{1-e^{-\lambda t_1}} \times \frac{1}{e^{-\lambda t_2}} \times \frac{\lambda t_3}{1-e^{-\lambda t_3}}$$

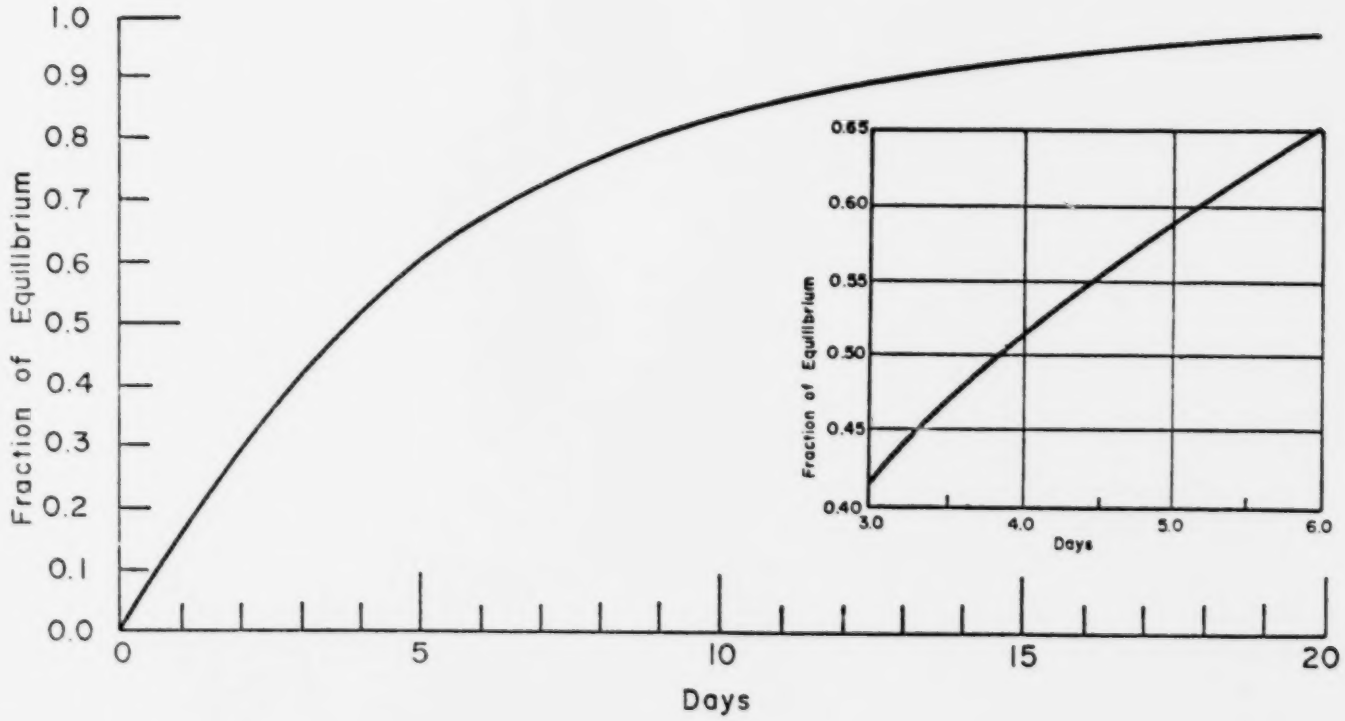

Figure 5. The Growth of Radon-222 from Radium-226

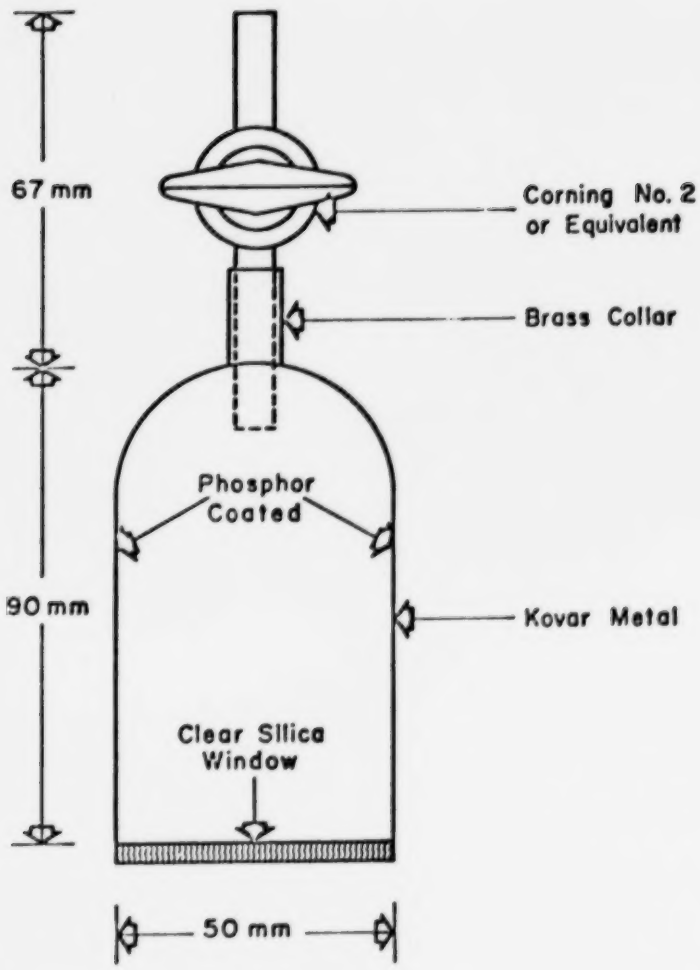

Figure 6. A Typical Scintillation Cell for Radon Counting

where:

C = net count rate, counts/minute (Note 4),
E = calibration constant for the de-emanation system and the scintillation cell in counts per minute/disintegrations per minute of ^{222}Rn (Note 5),
V = volume of sample in liters,
t_1 = the elapsed time in days between the first and second de-emanations (steps 6 and 12) and λ is the decay constant of ^{222}Rn (0.181 d^{-1}),
t_2 = the time interval between the second de-emanation and counting and λ is the decay constant of ^{222}Rn (0.00755 hr^{-1}), and
t_3 = the counting time in minutes and λ is the decay constant of ^{222}Rn (1.26 x 10^{-4} min^{-1}).

Notes:

1. The volume of these bubblers is usually greater than 20 ml allowing for at least a 1 cm air space between the bubbler and the stopper. In those instances where the solution volume exceeds the capacity of the bubbler, it will be necessary to continue the boiling in the water bath until the volume is reduced.

2. For minimizing corrections that would be required in subsequent calculations, the voids above the bubbler must be kept very small. Capillary tubing should be used whenever possible, and the drying tube volume with the Ascarite and magnesium perchlorate must be kept to a minimum. A typical system consists of a drying tube 10 cm x 1.0 cm (I.D.), with each of the drying agents occupying 4 cm and being separated by small glass wool plugs. The column can be reused several times before the chemicals need to be replaced.

3. After each analysis, flush the cell three times by evacuation and filling with helium, and store filled with helium at atmospheric pressure. This procedure removes radon from the cell and prevents the build-up of radon daughter products.

4. Before each analysis, the scintillation cell should be evacuated, filled with helium and counted to ascertain the cell background.

5. The calibration constant, E, is determined as follows:

   a. Place 50 pCi of the ^{226}Ra standard solution in a bubbler (50 pCi of ^{226}Ra will produce about 6 pCi ^{222}Rn in 18 hours). Attach the bubbler to the assembly as shown in Fig. 3.

   b. With the scintillation cell disconnected, bubble helium gas through the solution for 20 minutes.

   c. Close both stopcocks on the bubbler to establish zero time for ingrowth of ^{222}Rn. Set aside for approximately 18 hours.

   d. Evacuate the scintillation cell and attach to the column and bubbler.

   e. Proceed with steps 8-13, Radon Emanation Technique, p. 19.

   f. The calibration constant, E, is determined from the ^{226}Ra activity in the bubbler and the ingrowth time of ^{222}Rn by the equation:

$$E = \frac{C}{A\ (1-e^{-\lambda t_1})(e^{-\lambda t_2})}$$

where:

C = net count rate, counts/minute,
A = activity of ^{226}Ra in the bubbler (d/m),
t_1 = ingrowth time of ^{222}Rn in hours,
t_2 = decay time of ^{222}Rn in hours occurring between de-emanation and counting, and
λ = decay constant of ^{222}Rn, 0.00755 $hour^{-1}$.

The calibration constant, E, includes the de-emanation efficiency of the system, the counting efficiency of the cell, and the alpha activity contributed by ^{218}Po and ^{214}Po, which will be in equilibrium with ^{222}Rn when the sample is counted 4 hours after the de-emanation. A 100-minute counting time will be sufficient for the standard and will eliminate the need to correct for decay of ^{222}Rn, which occurs during counting.

The bubbler used for the ^{226}Ra standardization should not be used for sample analysis. It should be set aside to be retained for future calibrations. Each scintillation cell should be calibrated periodically with the ^{226}Ra standard to ensure instrument quality control.

Literature:

Blanchard, R. L. Uranium Decay Series Disequilibrium in Age Determination of Marine Calcium Carbonates. Doctoral Thesis, Washington University, St. Louis, Mo. (June 1963).

Ferri, E., P. J. Magno, and L. R. Setter. Radionuclide Analysis of Large Numbers of Food and Water Samples. U. S. Department of Health, Education, and Welfare, Public Health Service Publication No.999-RH-17 (1965).

Rushing, D. E. The Analysis of Effluents and Environmental Samples from Uranium Mills and of Biological Samples for Uranium, Radium and Polonium. SM/41-44, Symposium on Radiological Health and Safety, Vienna, Austria (August 1963).

RADIUM-228 IN DRINKING WATER

Sequential Method Radium-228/Radium-226

Principle of Method

The ^{228}Ra and ^{226}Ra in the drinking water sample are concentrated and separated by coprecipitation with barium and lead as sulfates and purified by EDTA-chelation. After 36-hour ingrowth of actinium-228 from radium-228, the ^{228}Ac is carried on yttrium oxalate, purified and beta counted. The radium-226 in the supernatant is either precipitated as the sulfate, purified and alpha counted (see step 20), or is transferred to a radon bubbler and determined by emanation (see step 27).

Procedure Time

Two samples in 12 hours.

Reagents

Acetic acid, $HC_2H_3O_2$: 17.4 N (glacial)
Acetone, $(CH_3)_2CO$: anhydrous
Ammonium hydroxide, NH_4OH: 15 N (conc.)
Ammonium oxalate, $(NH_4)_2C_2O_4.H_2O$: 5%
Ammonium sulfate, $(NH_4)_2SO_4$: 200 mg/ml
Ammonium sulfide, $(NH_4)_2S$: 2%
Barium carrier: 16 mg/ml
Citric acid, $C_6H_8O_7.H_2O$: 1 M
EDTA reagent: 0.25 M
Ethanol, C_2H_5OH: 95%
Indicator, methyl orange: 0.1%
Lead carrier: 15 mg/ml, 1.5 mg/ml
Nitric acid, HNO_3: 16 N (conc.), 6 N, 1 N
Sodium hydroxide, NaOH: 18 N, 10 N, 1 N
Strontium-yttrium mixed carrier: 0.9 mg/ml Sr^{+2}-0.9 mg/ml Y^{+3}
Sulfuric acid, H_2SO_4: 18 N
Yttrium carrier: 18 mg/ml, 9 mg/ml

Procedure

1. For each liter of drinking water, add 5 ml 1 M $C_6H_8O_7.H_2O$ and a few drops methyl orange indicator. The solution should be red (Note 1).
2. Add 10 ml lead carrier (15 mg/ml), 2.0 ml barium carrier (16 mg/ml), and 1 ml yttrium carrier (18 mg/ml); stir well. Heat to incipient boiling and maintain at this temperature for 30 minutes.
3. Add 15 N NH_4OH until a definite yellow color is obtained, then add a few drops excess. Precipitate lead and barium sulfates by adding 18 N

H_2SO_4 until the red color reappears, then add 0.25 ml excess. Add 5 ml $(NH_4)_2SO_4$ (200 mg/ml) for each liter of sample. Stir frequently and keep at a temperature of about 90°C for 30 minutes.

4. Cool slightly, then filter with suction through a 47-mm metricel membrane filter (GA-6,0.45 μ-pore size). Make a quantitative transfer of precipitate to the filter by rinsing last particles out of beaker with a strong jet of water.
5. Carefully place filter with precipitate in the bottom of a 250 ml beaker. Add about 10 ml 16 N HNO_3 and heat gently until the filter completely dissolves. Transfer the precipitate with the aid of more 16 N HNO_3 into a polypropylene centrifuge tube. Centrifuge and discard supernatant.
6. Wash the precipitate with 15 ml 16 N HNO_3, centrifuge, and discard supernatant.
7. Repeat step 6.
8. Add 25 ml EDTA reagent, heat in a hot water bath, and stir well. Add a few drops 10 N NaOH if the precipitate does not readily dissolve.
9. Add 1 ml strontium-yttrium mixed carrier and stir thoroughly. Add a few drops 10 N NaOH if any precipitate forms.
10. Add 1 ml $(NH_4)_2SO_4$ (200 mg/ml) and stir thoroughly. Add 17.4 N $HC_2H_3O_2$ until barium sulfate precipitates, then add 2 ml excess. Digest in a hot water bath until precipitate settles. Centrifuge and discard supernatant.
11. Add 20 ml EDTA reagent, heat in a hot water bath, and stir until precipitate dissolves. Repeat steps 9 and 10. (Note time of last barium sulfate precipitation; this is the beginning of the ^{228}Ac ingrowth time.)
12. Dissolve the precipitate in 20 ml EDTA reagent as before, then add 1.0 ml yttrium carrier (9 mg/ml) and 1 ml lead carrier (1.5 mg/ml). If any precipitate forms, dissolve by adding a few drops 10 N NaOH. Cap the polypropylene tube and age at least 36 hours.
13. Add 0.3 ml $(NH_4)_2S$ and stir well. Add 10 N NaOH dropwise with vigorous stirring until lead sulfide precipitates, then add 10 drops excess. Stir intermittently for about 10 minutes. Centrifuge and decant supernatant into a clean tube.
14. Add 1 ml lead carrier (1.5 mg/ml), 0.1 ml $(NH_4)_2S$, and a few drops 10 N

NaOH. Repeat precipitation of lead sulfide as before. Centrifuge and filter supernatant through Whatman #42 filter paper into a clean tube. Wash filter with a few ml water. Discard residue.

15. Add 5 ml 18 N NaOH (make at least 2 normal in OH^-). Stir well and digest in a hot water bath until yttrium hydroxide coagulates. Centrifuge and decant supernatant into a beaker. Cover beaker and save supernatant for ^{226}Ra analysis, step 20 or 27. (Note time of yttrium hydroxide precipitation; this is the end of the ^{228}Ac ingrowth time and beginning of ^{228}Ac decay time.)
16. Dissolve the precipitate in 2 ml 6 N HNO_3. Heat and stir in a hot water bath about 5 minutes. Add 5 ml water and reprecipitate yttrium hydroxide with 3 ml 10 N NaOH. Heat and stir in a hot water bath until precipitate coagulates. Centrifuge and discard supernatant.
17. Dissolve precipitate with 1 ml 1 N HNO_3 and heat in hot water bath a few minutes. Dilute to 5 ml and add 2 ml 5% $(NH_4)_2C_2O_4.H_2O$. Heat to coagulate, centrifuge and discard supernatant.
18. Add 10 ml water, 6 drops 1 N HNO_3 and 6 drops 5% $(NH_4)_2C_2O_4.H_2O$. Heat and stir in a hot water bath a few minutes. Centrifuge and discard supernatant.
19. Transfer quantitatively to a tared stainless-steel planchet with a minimum amount of water. Dry under an infra-red lamp to a constant weight and count in a low-background beta counter (Note 2).

Radium-226 - By Precipitation

20. To the supernatant from step 15, add 4 ml 16 N HNO_3 and 2 ml $(NH_4)_2SO_4$ (200 mg/ml), stirring well after each addition. Add 17.4 N $HC_2H_3O_2$ until barium sulfate precipitates, then add 2 ml excess. Digest on a hot plate until precipitate settles. Centrifuge and discard supernatant.
21. Add 20 ml EDTA reagent, heat in a hot water bath, and stir until precipitate dissolves. Add a few drops 10 N NaOH if precipitate does not readily dissolve
22. Add 1 ml strontium-yttrium mixed carrier, and 1 ml lead carrier (1.5 mg/ml); stir thoroughly. Add a few drops 10 N NaOH if any precipitate forms.
23. Add 1 ml $(NH_4)_2SO_4$ (200 mg/ml) and stir thoroughly. Add 17.4 N $HC_2H_3O_2$

until barium sulfate precipitates, then add 2 ml excess. Digest in a hot water bath until precipitate settles. Centrifuge and discard supernatant.

24. Wash precipitate with 10 ml water. Centrifuge and discard supernatant.
25. Transfer precipitate to a tared stainless-steel planchet with a minimum amount of water. Dry under an infra-red lamp and weigh (Note 3).
26. Count immediately in an alpha proportional counter.

Radium-226 - By Radon Emanation

27. To the supernatant from step 15, proceed with steps 20 to 25. Carefully transfer barium sulfate from planchet with the aid of a rubber policeman and 14 ml EDTA reagent into a small beaker. Add a few drops 10 N NaOH and heat to dissolve. Cool and transfer to a radon bubbler (Figure 4), rinsing beaker with another ml EDTA reagent, and proceed with Radium-226 in Drinking Water - Radon Emanation Technique, page 16, step 5.

Calculation for ^{228}Ra

Calculate the concentration, D, of ^{228}Ra in picocuries per liter as follows:

$$D = \frac{C}{2.22 \times EVR} \times \frac{\lambda t_2}{(1-e^{-\lambda t_2})^*} \times \frac{1}{(1-e^{-\lambda t_3})} \times \frac{1}{e^{-\lambda t_1}}$$

where:

C = average net count rate, counts/minute,
E = counter efficiency, for ^{228}Ac,
V = liters of sample used,
R = fractional chemical yield of yttrium carrier (step 19) multiplied by fractional chemical yield of barium carrier (step 25),
2.22 = conversion factor from disintegrations/minute to picocuries,
λ = the decay constant for ^{228}Ac (0.001884 min^{-1}),
t_1 = the time interval (in minutes) between the first yttrium hydroxide precipitation in step 15 and the start of the counting time,
t_2 = the time interval of counting in minutes, and
t_3 = the ingrowth time of ^{228}Ac in minutes measured from the last barium sulfate precipitation in step 11 to the first yttrium hydroxide precipitation in step 15.

* $\frac{\lambda t_2}{(1-e^{-\lambda t_2})}$ is a factor to correct the average count rate to count rate at beginning of counting time.

Calculation for ^{226}Ra

a) by precipitation, refer to Radium-226 in Drinking Water - Precipitation Method, page 14, b) by radon emanation, refer to Radium-226 in Drinking Water - Radon Emanation Technique, page 19.

Notes:

1. At the time of sample collection add 2 ml 16 N HNO_3 for each liter of water.
2. If the ^{226}Ra analysis is not desired, complete step 20 and then steps 24 and 25 to obtain the fractional barium yield for calculating ^{228}Ra activity.
3. If after sufficient beta decay of the actinium fraction, it is determined that there is no ^{228}Ra in the sample, then the ^{226}Ra fraction may be alpha counted directly. If ^{228}Ra is present, then the ^{226}Ra must be determined by radon emanation.

Literature:

Johnson, J. O. Determination of Radium-228 in Natural Waters. Radiochemical Analysis of Water, Geological Survey Water - Supply Paper 1696-G., U. S. Govt. Printing Office, Washington, D. C. (1971).

RADIOACTIVE STRONTIUM IN DRINKING WATER

Principle of Method

Strontium carrier is added to the drinking water sample, collected as the soluble carbonate, and separated from most of the calcium as the nitrate. Impurities are removed by an hydroxide scavenge. After the barium is removed as the chromate, the strontium is purified as $SrCO_3$ for counting.

Procedure Time

Four samples in 6 hours.

Reagents

Ammonium acetate buffer, (CH_3COOH-CH_3COONH_4): pH 5.0
Ammonium hydroxide, NH_4OH: 15 N (conc.), 6 N
Barium carrier: 16 mg/ml
Ethanol, C_2H_5OH: 95%
Hydrochloric acid, HCl: 1 N
Indicator, methyl red: 0.1%
Iron chloride, $FeCl_3$: 0.1 M
Nitric acid, HNO_3: 16 N (conc.), 6 N, 1 N
Oxalic acid, $H_2C_2O_4$: saturated
Sodium carbonate, Na_2CO_3: 1.5 M
Sodium chromate, Na_2CrO_4: 0.5 M
Sodium hydroxide, NaOH: 6 N
Strontium carrier: 20 mg/ml
Yttrium carrier: 10 mg/ml

Procedure

1. To a 1000-ml drinking water sample, add 1.0 ml strontium carrier and 1 ml barium carrier.
2. Make basic with 5 to 10 ml 6 N NaOH and heat to boiling.
3. Add 5 ml 1.5 M Na_2CO_3, stir, and digest until $SrCO_3$ coagulates, cool, centrifuge, and discard supernatant.
4. Wash precipitate with 15 ml water and discard wash solution.
5. Dissolve precipitate with 1 ml 6 N HNO_3.
6. Add 25 ml 16 N HNO_3, stir, and cool in an ice bath 5 minutes.
7. Centrifuge, discard supernatant, and let drain a few minutes (Note 1).
8. Dissolve precipitate with 10 ml water and add 0.5 ml 0.1 M $FeCl_3$.
9. Heat to near boiling in water bath and add 6 N NH_4OH dropwise until $Fe(OH)_3$ precipitates.

10. Cool, centrifuge, and transfer supernatant to a clean centrifuge tube. Discard precipitate. Note time of last precipitation; this is the beginning of yttrium ingrowth. (Complete steps 11 through 18 without delay to minimize ingrowth of ^{90}Y.)
11. Add 3 drops methyl red indicator, and adjust pH to near 5 with a few drops 1 N HCl. (Color change is from yellow to red.)
12. Add 5 ml ammonium acetate buffer solution and heat in water bath.
13. Slowly add 1 ml 0.5 M Na_2CrO_4. Stir, heat, and centrifuge. Transfer supernatant to a clean centrifuge tube; discard residue.
14. Add 2 ml 15 N NH_4OH to the supernatant, heat in water bath, and slowly add, with stirring, 5 ml 1.5 M Na_2CO_3. Digest until precipitation is complete, cool, centrifuge, and discard supernatant.
15. Dissolve precipitate with 5 ml 1 N HCl, add 10 ml water, and repeat step 14.
16. Wash the strontium carbonate precipitate with 20 ml water, and discard wash solution.
17. Slurry the precipitate with minimum of water and transfer to a tared stainless-steel planchet. Dry under infra-red lamps.
18. Cool, weigh, and beta count immediately (Note 2).

Calculation

Calculate the concentration, D, of the strontium activity in picocuries per liter as follows:

$$D = \frac{C}{2.22 \times EVR}$$

where:

C = net count rate, counts/minute,
E = counter efficiency,
V = liters of sample used,
R = fractional chemical yield, and
2.22 = conversion factor from disintegrations/minute to picocuries.

Notes:

1. If the drinking water samples contain much $CaCO_3$ (hardness), it will be necessary to repeat steps 5, 6, and 7 until it is all eliminated from the $Sr(NO_3)_2$ precipitate.
2. The counting result, immediately ascertained, represents the total strontium activity (^{90}Sr + ^{89}Sr) plus an insignificant fraction of the

^{90}Y that has grown in from the separated ^{90}Sr. To determine the ^{89}Sr and ^{90}Sr with a greater precision, the planchet should be stored at least 2 weeks so that the ^{90}Sr-^{90}Y activity will be in equilibrium. At this point, steps 19-39 are performed on the precipitate to separate the yttrium from the strontium and determine the ^{90}Sr activity.

19. After the period for ^{90}Y ingrowth, slurry the precipitate on the planchet with 2 ml water and transfer to a centrifuge tube with the aid of a rubber policeman. To make the transfer quantitative, wash the residue from the planchet with a small amount of 1 $\underline{N}$ HNO_3. Dissolve the precipitate in the tube with sufficient 1 $\underline{N}$ HNO_3, and dilute with water to 10 ml.
20. Add 1.0 ml yttrium carrier and stir.
21. Boil to expel dissolved carbon dioxide; cool to room temperature.
22. Replace in water bath and make basic with 2 to 3 ml 15 $\underline{N}$ NH_4OH. Stir and digest until the yttrium hydroxide precipitation is complete.
23. Cool, centrifuge, and decant supernatant into a 100-ml beaker. Note time of last precipitation; this is the end of ^{90}Y ingrowth and the beginning of ^{90}Y decay.
24. Dissolve precipitate in 1 ml 1 $\underline{N}$ HNO_3 and dilute with water to 10 ml.
25. Reprecipitate yttrium by dropwise addition of 15 $\underline{N}$ NH_4OH.
26. Centrifuge and combine supernatant with solution in the 100-ml beaker (step 23).
27. Repeat steps 24, 25, and 26. Save the combined supernatant solutions in the beaker for strontium activity and gravimetric yield determination, step 35 (Note 3).
28. Add 2 ml 1 $\underline{N}$ HNO_3 to the $Y(OH)_3$ precipitate from step 26 and dissolve. Dilute to 5 ml with water. Filter through Whatman #42 filter paper and collect filtrate in a centrifuge tube.
29. Slowly add 5 ml saturated $H_2C_2O_4$, with stirring, and digest in hot-water bath for 10 minutes.
30. Cool in an ice bath to room temperature.
31. Centrifuge and discard supernatant.
32. Wash precipitate twice with 10 ml hot water. Centrifuge and discard wash solutions.
33. Filter the yttrium oxalate on a tared glass-fiber filter. Wash with hot water and ethanol.

34. Dry, cool, weigh, mount, and beta count the ^{90}Y immediately.

35. Warm the combined supernatant solution from step 27, add 5 ml 1.5 M Na_2CO_3, and digest for 10 minutes.

36. Cool, centrifuge, and discard supernatant.

37. Wash the $SrCO_3$ with 15 ml water and discard wash solution.

38. Slurry with a few ml water and transfer quantitatively to a tared stainless-steel planchet. Dry under infra-red lamps.

39. Cool, weigh, and beta count immediately.

Calculation

Calculate the concentration of ^{89}Sr and ^{90}Sr in picocuries per liter as follows:

1. $$^{90}Y \text{ c/m (corrected)} = \frac{\text{c/m}}{A \times B_1 \times C}$$

2. $$^{90}Sr \text{ c/m} = {}^{90}Y \text{ c/m (corrected)} \times \frac{D}{E}$$

3. $$^{90}Y \text{ c/m} = \frac{^{90}Y \text{ c/m (corrected)} \times G \times B_2}{E}$$

4. $$^{89}Sr \text{ c/m} = (R - {}^{90}Sr \text{ c/m} - {}^{90}Y \text{ c/m}) \frac{1}{F}$$

5. $$^{90}Sr \text{ activity} = \frac{^{90}Y \text{ c/m (corrected)}}{2.22 \times EIV}$$

6. $$^{89}Sr \text{ activity} = \frac{^{89}Sr \text{ c/m}}{2.22 \times HIV}$$

where:

A = decay factor for ^{90}Y from step 23 to counting time,
B_1 = ingrowth factor of ^{90}Y from time of strontium purification to yttrium separation,
B_2 = ingrowth factor of ^{90}Y from time of yttrium separation to time of total strontium count,
C = fractional chemical yield for yttrium,
D = ^{90}Sr efficiency for counter in which radiostrontium is counted,
E = ^{90}Y efficiency for counter in which ^{90}Y is counted,
F = decay factor for ^{89}Sr from sample collection to counting time,
G = ^{90}Y efficiency for counter in which radiostrontium is counted,
H = ^{89}Sr efficiency for counter in which radiostrontium is counted,
I = fractional chemical yield for strontium,
R = observed count rate of total radiostrontium fraction (steps 18 or 39),
V = liters of sample used, and
2.22 = conversion factor from disintegrations/minute to picocuries.

Note:

3. Steps 35-39 are a repeat of the strontium carbonate precipitation to determine chemical yield after the yttrium has been removed. The beta activity should be comparable to that obtained previously for the precipitate from step 18. It is a more accurate result, however, since the only correction that need be made is that for the ingrowth of ^{90}Y from the time of yttrium separation (step 27) to the time of the total strontium count.

Literature:

Douglas, G. S., ed. Radioassay Procedures for Environmental Samples, Environmental Health Series, USDHEW Rept. 999-RH-27, National Center for Radiological Health, Rockville, Md. 10852 (Jan. 1967).

Hahn, R. B. and C. P. Straub. Determination of Radioactive Strontium and Barium in Water. J. Am. Water Works Assoc. 47 (4) 335-340 (April 1955).

TRITIUM IN DRINKING WATER

Principle of Method

The aqueous sample is distilled to dryness to effect quantitative transfer of tritium to the distillate and to remove interfering radionuclides and quenching materials. A portion of the distillate is mixed with scintillation solution and counted in a liquid scintillation spectrometer. Standard tritium and background samples are prepared and counted alternately to nullify errors produced by aging of the scintillation medium or instrument drift.

Procedure Time

Four samples in 2 hours.

Reagents

Scintillation solution: prepared reagent
Tritium, ^{3}H: standard tracer solution

Procedure

1. Distill the drinking water sample (50 ml or less) to dryness (Fig. 7) and collect the distillate in a centrifuge tube (Note 1).
2. Transfer 16 ml scintillation solution to a 25-ml scintillation vial.
3. Add 4 ml sample distillate to the scintillation vial, cap tightly, and shake until thoroughly mixed.
4. Prepare a background sample consisting of 4 ml water of minimal tritium concentration and 16 ml scintillation solution in same manner as sample.
5. Prepare a standard consisting of 16 ml scintillation solution and 4 ml water, which contains a standard concentration of tritium activity in same manner as sample.
6. Dark-adapt and cool sample, background, and standard solutions in instrument freezer to prepare for counting (Note 2).
7. In normal counting operation tritium is counted with a window setting where the figure of merit is at maximum (Note 3). The high voltage is set to obtain the peak counting efficiency in the window.
   To confirm the purity and identify the tritium activity the following steps are necessary:
8. Determine the count rate for each sample, background and standard. Three successive results which are within 2 sigma of each other, ensure that

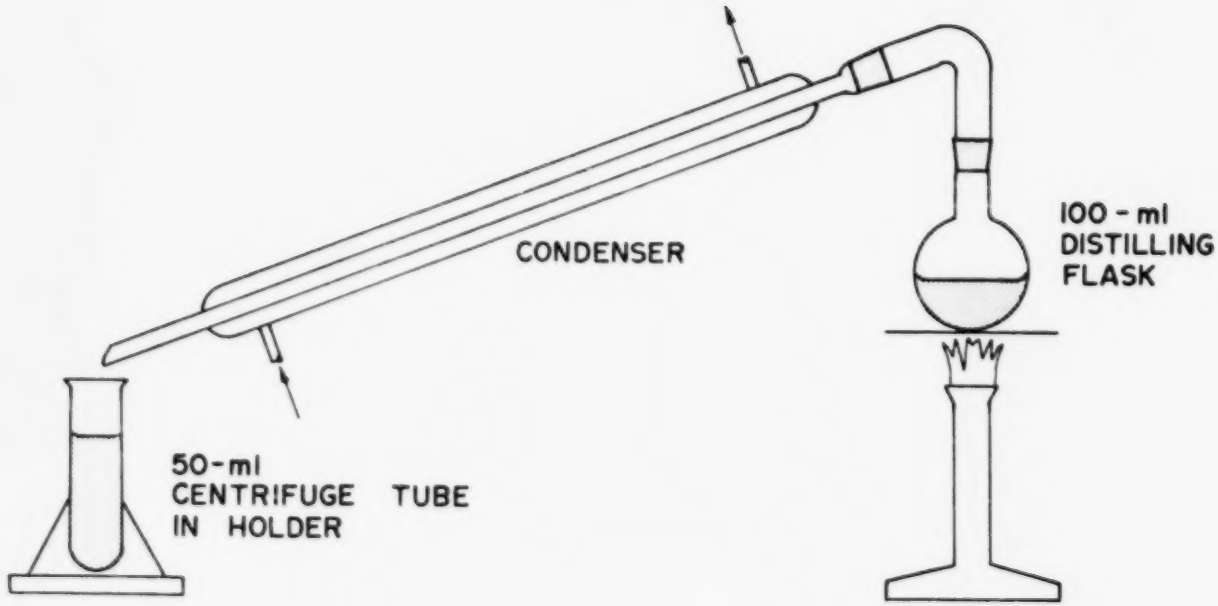

Figure 7. Distillation Apparatus for Tritium Analysis

the vials have been dark adapted.

9. With a 2-channel spectrometer, one window is set to give the best figure of merit and the other is set as an impurity screen. The ratios of the activity for the ^{3}H standard and for the distilled samples shoald be the same at the two window settings. If not, the samples must be redistilled to remove interfering ionizing radiations and prepared again for counting.

Calculation

Calculate the concentration, D, of the tritium activity in picocuries per liter as follows:

$$D = \frac{C \times 1000}{2.22 \times EV}$$

where:

C = net count rate, counts/minute,
E = efficiency for measuring ^{3}H in liquid scintillation spectrometer,
V = milliliters of sample used,
2.22 = conversion factor from disintegrations/minute to picocuries.

Calculate the efficiency, E, for measuring ^{3}H in the liquid scintillation spectrometer as follows:

$$E = \frac{Y}{S}$$

where:

Y = counts/minute determined by counting standard tritium sample (step 5) at the optimum instrument settings, and
S = standard tritium activity (disintegrations per minute/ml) as rated by NBS or equivalent, corrected for decay.

Calculate the decay correction for the tritium activity as follows:

$$A = A_{o}\, e^{-0.693t/T}$$

where:

A = activity at time t,
A_o = activity at time of collection or from the date the standard was rated,
e = base of natural logarithms,
t = elapsed time from collection or from the date the standard was rated, and
T = half life of tritium (12.3 years).

Notes:

1. As a general rule, all samples should be distilled to dryness for quantitative recovery of tritium and to remove interfering radionuclides. Iodine-131 in aqueous samples can be eliminated by adding stable iodine and $AgNO_3$ to the flask before the distillation.
2. The freezer temperature must be maintained above $2^{o}C$ or, in time, the solution will begin to solidify. If an ambient temperature liquid scintillation spectrometer is employed, the vial must be dark-adapted, usually 24 to 48 hours, before counting begins.
3. Figure of Merit = $\frac{E^2 \ (\text{Efficiency})^2}{B \quad (\text{Background})}$

Literature:

Butler, F. E. Determination of Tritium in Water and Urine. Anal. Chem. 33, 409-414 (1961).

POTASSIUM-40 IN DRINKING WATER (Note 1)

Principle of Method

Total potassium in drinking water is determined by one of several methods--atomic absorption, flame photometry, or colorimetry. From the determination of mg/l total potassium present, the concentration of ^{40}K can be calculated.

Preparation of Standards

Stock Solution: Dissolve 0.1907 g of KCl (analytical reagent grade), dried at 110°C, in deionized distilled water and make up to 1 liter.
1 ml = 0.10 mg K (100 mg/l).

Prepare dilutions of the stock solution to be used as calibration standards at the time of analysis.

General Instrumental Parameters for Atomic Absorption (Note 2)

Potassium hollow cathode lamp
Wavelength: 766.6 nm
Fuel: Acetylene
Oxidant: Air
Type of flame: Slightly oxidizing

Procedure

1. For determining total potassium, the drinking water sample is acidified with 1:1 redistilled HNO_3 to a pH of 2 at the time of collection. The sample is not filtered before processing.
2. Transfer a representative aliquot of the drinking water sample (50 to 100 ml) to a Griffin beaker and add 3 ml of concentrated redistilled HNO_3. Place the beaker on a hot plate and evaporate to dryness cautiously, making certain that the sample does not boil. Cool the beaker and add another 3 ml portion of concentrated redistilled HNO_3. Cover the beaker with a watch glass and return to the hot plate. Increase the temperature of the hot plate so that a gentle reflux action occurs. Continue heating, adding additional acid as necessary, until the digestion is complete (generally indicated by a light colored residue). Add sufficient distilled 1:1 HCl and again warm the beaker to dissolve the residue.

3. Wash down the beaker walls and watch glass with distilled water and filter the sample to remove silicates and other insoluble material that could clog the atomizer. Adjust the volume to some predetermined value based on the expected potassium concentration. The sample is now ready for analysis (Notes 3 and 4).

Calculation

Calculate the concentration, D, of ^{40}K in drinking water in picocuries per liter as follows:

$$D = \frac{K \times 1.883}{2.22}$$

where:

K = concentration of potassium in mg/l of sample,
1.883 = disintegration/minute for each mg potassium,
2.22 = conversion factor from disintegrations/minute to picocuries.

Notes:

1. This technique[1] is presented for those interested in calculating the ^{40}K in drinking water. All that is required is that total potassium be determined in any manner available, and from this value, the radioactive potassium value can be calculated.
2. The Osram potassium vapor-discharge lamp may also be used in the Perkin-Elmer 303. In this case, the current should be 350 ma or the optimum operating current.
   a. Sodium may interfere if present at much higher levels than the potassium. This effect can be compensated by approximately matching the sodium content of the potassium standards with that of the sample.
   b. Potassium absorption is enhanced in the presence of Na, Li, and Cs, especially in a high-temperature flame. This enhancement effect of sodium can be eliminated by changing the burner height and the type of flame used. The burner assembly is set approximately 0.05 cm below the optical light path so that the optical light path is sliced at the bottom by the burner head. A fuel-rich flame is used.
   c. The 404.4 nm line may also be used. This line has a sensitivity of 5 mg/l for 1% absorption.
   d. To cover the range of potassium values normally observed in surface waters (0.1 to 20 mg/l), it is suggested that the burner be rotated 75°.
3. Flame photometric or colorimetric methods[2] may be used if atomic absorption instruments are not available.

4. The precision and accuracy of the technique has been determined at the Methods Development and Quality Assurance Research Laboratory (MDQARL). Using distilled water samples at concentrations of 1.6 and 6.3 mg/l, the standard deviations were ± 0.2 and ± 0.5, respectively. Recoveries at these levels were 103% and 102%. With an optimum concentration range between 0.1 and 2 mg/l and with the use of a wavelength of 766.5 nm, the sensitivity was 0.04 mg/l and the detection limit was 0.005 mg/l.

References:

1. Methods for Chemical Analysis of Water and Wastes, p. 143. Methods Development and Quality Assurance Research Laboratory, National Environmental Research Center-Cincinnati, EPA-625/6-74-003 (1974).

2. Standard Methods for the Examination of Water and Waste Water, 13th ed. pp. 283-285. American Public Health Association, Washington, D. C. (1971).

APPENDIX A

METHOD CAPABILITIES

METHOD CAPABILITIES*

	Parameter			3σ MDL pCi/l	
	Sample Volume	Counting Efficiency %	Instrument Background	1000 min count	60 min count
Gross Beta	100 ml	39	1.0 c/m	1.5	6
Gross Alpha	100 ml	41	0.2	0.8	2.4
^{134}Cs	1000 ml	31	1.2	0.2	0.8
^{137}Cs	1000 ml	34	1.2	0.2	0.8
^{131}I	2000 ml	32	1.0	0.1	0.4
^{226}Ra-precipitation	2000 ml	40	0.5	0.05	0.2
^{226}Ra-Rn emanation	1000 ml	70	0.03	0.02	0.06
^{228}Ra	2000 ml	40	1.5	0.1	0.4
^{89}Sr	1000 ml	42	1.2	0.3	1.3
^{90}Sr	1000 ml	33	1.2	0.2	0.8
Tritium	4 ml	30	9.0	200	600

* With these parameters, the procedures in this manual can obtain the corresponding minimum detection levels (MDL), assuming good chemical yield recovery. The calculations were made with data from replicate testing of these procedures. Variability in instrument background is a significant factor in the determination of these values. The levels reported above can be further reduced by the use of larger samples for analysis and better instrument shielding.

APPENDIX B

REAGENT PREPARATION

Distilled or deionized water should be used to prepare all reagents requiring water as the solvent.

I. Carrier solutions - These solutions, prepared as the specific ion, are to be filtered and standardized before use in chemical yield determinations. Reagent blanks should be prepared in the appropriate geometry with approximate final precipitate weight to ascertain instrument plus reagent background.

Ba^{++} - 16 mg/ml. Dissolve 2.846 grams $BaCl_2.2H_2O$ in water, add 0.5 ml 16 N HNO_3, and dilute to 100 ml with water.

Cs^{+} - 10 mg/ml. Dissolve 1.267 grams CsCl in water and dilute to 100 ml.

I^{-} - 20 mg/ml. Dissolve 2.616 grams KI in water, add 2 drops Na_2SO_3, and dilute to 100 ml. Store in dark flask.

IO_3^{-} - 10 mg/ml. Dissolve 1.685 grams KIO_3 in water and dilute to 100 ml. Store in dark flask.

Pb^{++} - 15 mg/ml. Dissolve 2.397 grams $Pb(NO_3)_2$ in water, add 0.5 ml 16 N HNO_3, and dilute to 100 ml with water.

Pb^{++} - 1.5 mg/ml. Dilute 10.0 ml $Pb(NO_3)_2$ (15 mg/ml) to 100 ml with water.

Sr^{++} - 20 mg/ml. Dissolve 4.831 grams $Sr(NO_3)_2$ in water and dilute to 100 ml.

Y^{+3} - 18 mg/ml. Add 22.85 grams Y_2O_3 to an Erlenmeyer flask containing 20 ml water. Heat to boiling and continue stirring with a magnetic stirring hot plate while adding 16 N HNO_3 in small amounts. Usually about 30 ml 16 N HNO_3 is necessary to dissolve the Y_2O_3. Small additions of water may be required to replace that lost by evaporation. After total dissolution add 70 ml 16 N HNO_3 and dilute to 1 liter with water.

Y^{+3} - 10 mg/ml. Dissolve 43.1 grams $Y(NO_3)_3.6H_2O$ in 800 ml water, add 5 ml 6 N HNO_3, and dilute to 1 liter.

Sr^{++} - Y^{+3} (mixed carrier) - (0.9 mg/ml Sr^{+2} and ~0.9 mg/ml Y^{+3}).

Solution A - Dilute 10.0 ml yttrium carrier Y^{+3} (18 mg/ml) to 100 ml.

Solution B - Dissolve 0.4348 grams $Sr(NO_3)_2$ in water and dilute to 100 ml. Combine Solutions A and B and label.

II. Acids and Inorganic Reagents

Ammonium acetate buffer, (CH_3COOH-CH_3COONH_4): pH 5.0. Mix 100 ml 1.5 N CH_3COOH and 100 ml 3 M CH_3COONH_4.

Acetic acid, CH_3COOH, 17.4 N: This is the concentrated (glacial) reagent; sp. gr. 1.06, 99.5%.

Acetic acid, CH_3COOH, 1.5 N: Dilute 86 ml glacial (17.4 N) acetic acid to 800 ml with water and dilute to 1 liter.

Ammonium acetate, CH_3COONH_4, 3 M: Dissolve 231 grams CH_3COONH_4 in 600 ml water and dilute to 1 liter.

Ammonium hydroxide, NH_4OH, 15 N: This is the concentrated reagent; sp. gr. 0.9, 50%.

Ammonium hydroxide, 6 N: Add 400 ml 15 N NH_4OH to 400 ml water and dilute to 1 liter.

Ammonium hydroxide, 2 N: Dilute 100 ml 6 N NH_4OH to 300 ml with water.

Ammonium oxalate, $(NH_4)_2C_2O_4$, 5%: Dissolve 25 grams $(NH_4)_2C_2O_4$ in water and dilute to 500 ml.

Ammonium phosphomolybdate (prepared reagent): Dissolve 100 grams of molybdic acid (85% MoO_3) in a mixture of 240 ml of water and 140 ml 15 N NH_4OH. When solution is complete, filter and add 60 ml of 16 N HNO_3. Mix 400 ml of 16 N HNO_3 and 960 ml of water. Allow both solutions to cool to room temperature. With constant stirring, add the ammonium molybdate solution to the nitric acid solution. Allow to stand for 24 hours. Filter through Whatman #42 filter paper. Discard the insoluble material.

Collect the filtrates in a 3-liter beaker and heat to 50° to 55°C. Remove from heating unit. It is important that the solution not be heated above 55°C to avoid contamination of the precipitate with molybdic anhydride. Add 25 grams of NaH_2PO_4 dissolved in 100 ml of water to the ammonium molybdate solution. Stir occasionally for 15 minutes and allow the precipitate to settle (approximately 30 minutes). Filter through Whatman #42 filter paper. Wash the precipitate with 1% potassium nitrate and finally with water. Dry the precipitate and paper at 100°C for 3 to 4 hours. Transfer the $(NH_4)_3(PMo_{12}O_{40})$ solid to a weighing bottle, and store in a desiccator.

Ammonium sulfate, 200 mg/ml: Dissolve 20 grams $(NH_4)_2SO_4$ in a minimum of water and dilute to 100 ml.

Ammonium sulfide, 2%: Dilute 10 ml $(NH_4)_2S$, (20-24%), to 100 ml with water.

Calcium chloride, 3 M: Dissolve 330 grams $CaCl_2$ in water and dilute to 1 liter.

Chloroplatinic acid, 0.1 M: Dissolve 51.8 grams $H_2PtCl_6.6H_2O$ in water and dilute to 1 liter.

Citric acid, 1 M: Dissolve 19.2 grams $C_6H_8O_7$ in water and dilute to 100 ml.

Helium, gas

Hydrochloric acid, HCl, 12 N: This is the concentrated reagent; sp. gr. 1.19, 37%.

Hydrochloric acid, 6 N: Add 500 ml 12 N HCl to 400 ml water and dilute to 1 liter.

Hydrochloric acid, 4 N: Add 333 ml 12 N HCl to 500 ml water and dilute to 1 liter.

Hydrochloric acid, 2 N: Dilute 333 ml 6 N HCl to 1 liter with water.

Hydrochloric acid, 1 N: Dilute 250 ml 4 N HCl to 1 liter with water.

Hydrofluoric acid, HF, 48% (~ 30 N): This is the concentrated reagent; sp. gr. 1.15.

Iron chloride, 0.1 M: Dissolve 27 grams $FeCl_3.6H_2O$ in water plus 2 ml 12 N HCl and dilute to 1 liter.

Magnesium perchlorate, $Mg(ClO_4)_2$: reagent grade.

Nitric acid, HNO_3, 16 N: This is the concentrated reagent; sp. gr. 1.42, 70%.

Nitric acid, 6 N: Cautiously add 395 ml 16 N HNO_3 to 600 ml water and dilute to 1 liter.

Nitric acid, 4 N: Cautiously add 250 ml 16 N HNO_3 to 700 ml water and dilute to 1 liter.

Nitric acid, 1 N: Add 62 ml 16 N to HNO_3 to 900 ml water and dilute to 1 liter.

Nitric acid, 0.2 N: Add 12.5 ml 16 N HNO_3 to 900 ml water and dilute to 1 liter.

Oxalic acid, saturated: Dissolve 150 grams $H_2C_2O_4$ in 1 liter boiling water.

Palladium chloride, 0.2 M: Dilute 118 ml 5% $PdCl_2$ to 167 ml with 2 N HCl.

Phosphoric acid, H_3PO_4, 85% (44 N): This is the concentrated reagent; sp. gr. 1.69.

Silver nitrate, 0.1 M: Dissolve 17 grams $AgNO_3$ in water and dilute to 1 liter. Store in dark flask.

Sodium bisulfite, 1 M: Dissolve 5.2 grams $NaHSO_3$ in water and dilute to 50 ml. Prepare only in small quantities.

Sodium carbonate, 1.5 M: Dissolve 160 grams Na_2CO_3 in 600 ml water and dilute to 1 liter.

Sodium chromate, 0.5 M: Dissolve 171.1 grams $Na_2CrO_4.10H_2O$ in 400 ml water and dilute to 1 liter.

Sodium hydroxide, 18 N: Dissolve 720 grams NaOH in 500 ml water and dilute to 1 liter.

Sodium hydroxide, 10 N: Dissolve 400 grams NaOH in 500 ml water and dilute to 1 liter.

Sodium hydroxide, 6 N: Dissolve 240 grams NaOH in 800 ml water and dilute to 1 liter.

Sodium hydroxide, 1 N: Dilute 100 ml 10 N NaOH to 1 liter with water.

Sodium hydroxide, 0.5 N: Dilute 50 ml 6 N NaOH to 600 ml with water.

Sodium nitrite, 1 M: Dissolve 69 grams $NaNO_2$ in water and dilute to 1 liter.

Sodium sulfite, 1 M: Dissolve 5 grams Na_2SO_3 in 40 ml water. Prepare fresh reagent every week.

Sulfuric acid, H_2SO_4, 36 N: This is the concentrated reagent; sp. gr. 1.84, 95-98%.

Sulfuric acid, 18 N: Cautiously add, with stirring, 500 ml 36 N H_2SO_4 to 400 ml water and dilute to 1 liter.

Sulfuric acid, 12 N: Cautiously add, with stirring, 333 ml 36 N H_2SO_4 to 500 ml water and dilute to 1 liter.

Sulfuric acid, 9 N: Cautiously add, with stirring, 250 ml 36 N H_2SO_4 to 600 ml water and dilute to 1 liter.

Sulfuric acid, 2 N: Dilute 100 ml 12 N H_2SO_4 to 600 ml with water.

Sulfuric acid, 0.1 N: Dilute 50 ml 2 N H_2SO_4 to 1 liter with water.

Zinc, powder: Reagent grade.

III. Organic reagents

Acetone, $(CH_3)_2CO$, anhydrous

Ascarite, granular, 8 to 20 mesh

Carbon tetrachloride, CCl_4

Diethyl ether, $(C_2H_5)_2O$, anhydrous

EDTA reagent: Dissolve 20 grams NaOH in about 750 ml water, heat, and slowly add 93 grams $Na_2C_{10}H_{14}O_8N_2.2H_2O$ (disodium ethylenedinitriloacetate dihydrate) while stirring. After the salt is in solution, filter through coarse filter paper and dilute to 1 liter. This solution is ~ 0.25 M.

Ethanol, C_2H_5OH, 95%

Scintillation solution*: (If prepared in the presence of daylight or fluorescent light, store in dark place 2 days before use.)

Dissolve 120 grams naphthalene, 0.05 grams 1,4-di (2-(5-phenyloxazolyl) benzene) (POPOP), and 4 grams 2, 5-diphenyloxazole (PPO), in 1 liter of p-dioxane. Store in an amber-colored bottle. Scintillation grade reagents are recommended to ensure sample stability.

Tartaric acid, 50%: Dissolve 50 grams $C_4H_6O_6$ in water and dilute to 100 ml.

IV. Indicators

Methyl red, 0.1%: Dissolve 0.1 grams methyl red indicator in 100 ml ethanol.

Methyl orange, 0.1%: Dissolve 0.1 grams methyl orange indicator in 100 ml water.

* Commercially prepared scintillation solutions are available from several supply houses, and can be substituted for this prepared stock solution.

APPENDIX C

SOURCES OF SUPPLY

Glass-fiber filter paper:

Reeve Angel
9 Bridewell Place
Clifton, N. J.

Specification: 2.8 cm Grade 934 AH

Liquid scintillation vials:

Packard Instrument Co.
2200 Warrenville Rd.
Downers Grove, Ill. 60515

Catalog #6001075 Polyethylene vial 20 ml, w/22 mm screw cap

Membrane filters:

Gelman Instrument Co.
Ann Arbor, Mich. 48106

Specification: Metricel, A-6, 47 mm, 0.45 μ

Mylar film:

Cadillac Plastics
3818 Red Bank Rd.
Cincinnati, Ohio 45227

Specification: 0.0005 inch thick

Plastic rings and discs:

Control Molding Corporation
84 Granite Ave.
Staten Island, N. Y. 10303

Catalog #J-356 1-inch dia (natural color) nylon type 6/6

Polypropylene centrifuge tube:

Dynalab Corp.
P. O. Box 112
Rochester, N. Y. 14601

Catalog #3103-0050 134 x 28.7 mm O.D. Closure #29C

Radon counter:

Randam Electronics, Inc.
3091 Shadycrest Drive
Cincinnati, Ohio 45236

Johnson Laboratories, Inc.
3 Industry Lane
Cockeysville, Md. 21030

Specifications: Bias network, detector circuits, 6 digit decade counter. External high voltage supply and 2-inch photomultiplier tubes.

Radon emanation bubblers and associated glassware: These can be fabricated by local glass companies with the specifications from Figures 3 and 4.

Scintillation cell:

Johnson Laboratories, Inc.
3 Industry Lane
Cockeysville, Md. 21030

Specifications: according to Figure 6.

Stainless-steel planchets:

Hruden Laboratory Products
P. O. Box 1802
Ann Arbor, Mich. 48106

Catalog #75750 2 x 0.018 x 1/4 inches.

Teflon filter holder:

Atomic Products Corp.
Center Moriches, N. Y. 11934